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[Inventor]

[Address]

c/o Mitsui Chemicals, Inc.  
580-32, Nagaura, Sodegaura  
Chiba, Japan

[Name]

Hideyuki KANEKO

[Inventor]

[Address]

c/o Mitsui Chemicals, Inc.  
580-32, Nagaura, Sodegaura  
Chiba, Japan

[Name]

Shinichi KOJOH

[Inventor]

[Address]

c/o Mitsui Chemicals, Inc.  
580-32, Nagaura, Sodegaura  
Chiba, Japan

[Name]

Nobuo KAWAHARA

[Inventor]

[Address]

c/o Mitsui Chemicals, Inc.  
580-32, Nagaura, Sodegaura  
Chiba, Japan

[Name]

Shingo MATSUO

[Inventor]

[Address]

c/o Mitsui Chemicals, Inc.  
580-32, Nagaura, Sodegaura

	Chiba, Japan
[Name]	Tomoaki MATSUGI
[Inventor]	
[Address]	c/o Mitsui Chemicals, Inc. 580-32, Nagaura, Sodegaura Chiba, Japan
[Name]	Norio KASHIWA
[Applicant for Patent]	
[Identification No.]	000005887
[Name]	Mitsui Chemicals, Inc.
[Representative]	Hiroyuki NAKANISHI
[Designation of Charge]	
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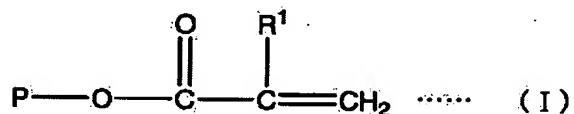
[Name of Document] SEPECIFICATION

[Title of Invention] POLYOLEFIN MACROMONOMER AND PROCESS OF PRODUCING THE SAME

[Claims]

[Claim 1] A polyolefin macromonomer having an acryloyl group or a methacryloyl group at the terminal of a polyolefin chain P, represented by the following general formula (I):

[Chemical Formula 1]

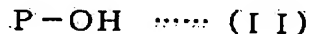


(wherein R<sup>1</sup> is a hydrogen atom or a methyl group, and P is a polyolefin chain obtained by homopolymerizing or copolymerizaing olefins represented by CH<sub>2</sub>=CHR<sup>2</sup> (where R<sup>2</sup> is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom).)

[Claim 2] A process of producing the polyolefin macromonomer according to Claim 1 which is obtained by successively carrying out the following steps (A) and (B):

Step (A): producing polyolefin having a hydroxyl group at the terminal of a polyolefin chain P, represented by the following general formula (II):

[Chemical Formula 2]



(wherein P has the same meaning as defined in the general formula (I).)

Step (B): converting a terminal hydroxyl group in the polyolefin chain P obtained in the step (A) into an acryloyl group or a methacryloyl group.

[Claim 3] The polyolefin macromonomer according to Claim 1 or Claim 2, wherein the molecular weight distribution (Mw/Mn) of the polyolefin chain P is 1.5 or more.

[Detailed Description for the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to a polyolefin macromonomer having an acryloyl group or a methacryloyl group at the terminal thereof, and a process of producing the same.

[0002]

[Description of the Related Art]

Polyolefin such as polyethylene (PE) and polypropylene (PP) are characterized by not only being lightweight and inexpensive but also having excellent physical properties and processability. But the high chemical stability of polyolefin gets in the way in terms of high functionality such as printability, coating ability, heat resistance, impact resistance, and compatibility with other polar polymers. As a method of compensating for this disadvantage

and endowing the polyolefin with above-described functionality, there is known, for example, a method of copolymerizing an olefin with a polar monomer such as vinylacetate or methacrylate by radical polymerization, or a method of grafting a polar monomer such as maleic anhydride onto polyolefin in the presence of a peroxide. However, in these methods, it is difficult to accurately regulate the structure of a polyolefin moiety in the resulting polymer, and thus not adequate to maintain excellent physical properties inherent in the polyolefin.

[0003]

As one means of producing a polymer having a polyolefin moiety whose structure is accurately regulated and having functions not exhibited by polyolefin only, there can be considered a method wherein a polyolefin macromonomer having a polymerizable vinyl bond at the terminal thereof is homopolymerized or is copolymerized with various vinyl monomers having functional groups to convert the monomer into a graft polymer having polyolefin side chains. As the method of producing a polyolefin macromonomer for synthesis of the graft polymer, for example, Japanese Unexamined Patent Application Publication No.6-329720 describes a method wherein a polymerizable acryloyl or methacryloyl group is introduced into the terminal of polyethylene synthesized by living polymerization. Japanese Unexamined

Patent Application Publication No.8-176354 and Japanese Unexamined Patent Application Publication No.8-176415 disclose a method of introducing a polymerizable styryl group into the terminal of a polypropylene polymer synthesized by living polymerization.

[0004]

In the methods using living polymerization, only one polymer is obtained from one active site on catalyst, and thus the molecular weight distribution ( $M_w/M_n$ ) of polyolefin is about 1. However, from the viewpoint of productivity, it is preferable that the number of polymers obtained from one active site on catalyst is larger, and from the viewpoint of moldability of the polymer, that the molecular weight distribution ( $M_w/M_n$ ) of polyolefin is greater. Accordingly, from economical viewpoint, the method utilizing the living polymerization is inadequate for industrial mass production of polyolefin. Further, the method described in Japanese Unexamined Patent Application Publication No.6-329720 utilizes anion polymerization using alkyl lithium, and thus polyolefin producible as a macromonomer is polyethylene having a relatively low molecular weight of a 1000-mer at the maximum. The method described in Japanese Unexamined Patent Application Publication No.8-176354 utilizes a vanadium compound having a specific structure as an olefin polymerization catalyst, and thus the type of producible

polyolefin is limited to a polypropylene polymer.

[0005]

[Problems to be Solved by the Invention]

Under these circumstances, the present inventors have made extensive and intensive studies for developing a polyolefin macromonomer which has excellent productivity and consists of various  $\alpha$ -olefin homopolymers or copolymers and is capable of covering a broad molecular weight range. As a result, they have found out a method wherein a hydroxyl group is introduced into the terminal of polyolefin produced by using a coordination polymerization catalyst containing a transition metal compound which is represented by a solid-phase titanium catalyst or a metallocene catalyst used widely in the industry as a catalyst for production of polyolefin, and an acryloyl group or a methacryloyl group is then further introduced into the polyolefin. These findings have led to the invention of a process of producing a novel polyolefin macromonomer by using an industrially useful method.

[0006]

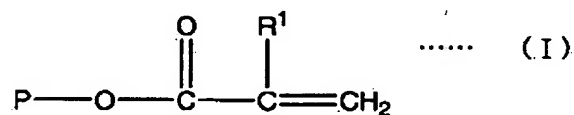
[Means for Solving the Problems]

Hereinafter, the process of producing a polyolefin macromonomer according to the present invention will be described in detail. The polyolefin macromonomer having an

acryloyl group or a methacryloyl group at the terminal of a polyolefin chain P according to the present invention, which is represented by the following general formula (I):

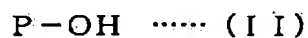
[0007]

[Chemical Formula 3]



(wherein R<sup>1</sup> is a hydrogen atom or a methyl group, and P is a polyolefin chain obtained by homopolymerizing or copolymerizing olefins represented by CH<sub>2</sub>=CHR<sup>2</sup> (where R<sup>2</sup> is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom).), can be produced by successively carrying out a step (A) of producing polyolefin having a hydroxyl group at the terminal of a polyolefin chain P represented by the following general formula (II):

[Chemical Formula 4]



(wherein P has the same meaning as defined in the general formula (I).), and a step (B) of converting a terminal hydroxyl group in the polyolefin chain P obtained in the step (A) into an acryloyl group or a methacryloyl group.

[0009]

Production of polyolefin having a hydroxyl group at the



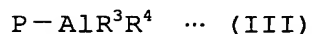
terminal thereof

The polyolefin having a hydroxyl group at the terminal of polyolefin chain P can be produced, for example, by producing polyolefin modified at the terminal thereof in the presence of an olefin polymerization catalyst, then subjecting a terminal group in the polyolefin modified at the terminal thereof to substitution reaction with a compound having a functional group structure, and subjecting the product to solvolysis, or subjecting a terminal group in the polyolefin modified at the terminal thereof to substitution reaction with a compound giving a structure which forms a functional group by solvolysis, and subjecting the product to solvolysis.

Hereinafter, the present invention comprising a step of producing polyolefin modified at the terminal thereof and a step of converting the polyolefin modified at the terminal thereof into polyolefin (II) having a hydroxyl group at the terminal will be successively described in detail.

< Production of polyolefin modified at the terminal thereof >

The polyolefin modified at the terminal thereof have, for example, the following general formula (II):



(wherein P is a polyolefin chain obtained by

homopolymerizing or copolymerizaing olefins represented by  $\text{CH}_2=\text{CHR}^2$ ,  $\text{R}^3$  and  $\text{R}^4$  may be the same or different from each other and each is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom, and  $\text{R}^2$  is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom.)

First, the olefin polymerization catalyst used in the production of polyolefin modified at the terminal thereof represented by the general formula (III) is described. The olefin polymerization catalyst used in the production of polyolefin modified at the terminal thereof may be any catalyst known in the art. The catalyst known in the art includes, for example, a magnesium-supported titanium catalyst, a metallocene catalyst, a post-metallocene catalyst, and the like. Hereinafter, these catalysts are described.

[0010]

< Magnesium-supported titanium catalyst >

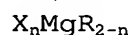
As an example of the catalyst used in the present invention, a polymerization catalyst composed of a solid titanium catalyst component (a) and an organometallic compound catalyst component (b) is described below.

[0011]

[Solid titanium catalyst (a)]

The solid titanium catalyst component (a) forming an olefin polymerization catalyst can be prepared by bringing a magnesium compound, a titanium compound, and an electron donor (i) into contact with each other as described below.

The magnesium compound includes a magnesium compound having reducing ability and a magnesium compound having no reducing ability. The magnesium compound having reducing ability includes, for example, an organomagnesium compound represented by the following formula:



(wherein  $n$  is a number of  $0 \leq n < 2$ ,  $R$  is hydrogen or an alkyl group, an aryl group, or a cycloalkyl group having 1 to 20 carbon atoms; when  $n$  is 0, two of  $R$ 's may be the same or different; and  $X$  is halogen). Examples of the organomagnesium compound having reducing ability include, in particular, alkyl magnesium compounds such as dimethyl magnesium, diethyl magnesium, dipropyl magnesium, dibutyl magnesium, diamyl magnesium, dihexyl magnesium, didecyl magnesium, octylbutyl magnesium, and ethylbutyl magnesium; alkyl magnesium halides such as ethyl magnesium chloride, propyl magnesium chloride, butyl magnesium chloride, hexyl magnesium chloride, and amyl magnesium chloride; alkyl magnesium alkoxides such as butylethoxy magnesium, ethylbutoxy magnesium, and octylbutoxy magnesium; butyl

magnesium hydride, and magnesium hydride.

[0012]

In addition, metallic magnesium can be used. Examples of the magnesium compounds having no reducing ability include, in particular, magnesium halides such as magnesium chloride, magnesium bromide, magnesium iodide, and magnesium fluoride; alkoxy magnesium halides such as methoxy magnesium chloride, ethoxy magnesium chloride, isopropoxy magnesium chloride, butoxy magnesium chloride, and octoxy magnesium chloride; aryloxy magnesium halides such as phenoxy magnesium chloride and methyl phenoxy magnesium chloride; dialkoxy magnesiums such as diethoxy magnesium, diisopropoxy magnesium, dibutoxy magnesium, di-n-octoxy magnesium, di-2-ethylhexoxy magnesium, and methoxyethoxy magnesium; diaryloxy dimagnesiums such as diphenoxy magnesium, di-methylphenoxy magnesium, and phenoxy-methylphenoxy magnesium; and magnesium carboxylates such as magnesium laurate and magnesium stearate.

[0013]

These magnesium compounds having no reducing ability may be compounds derived from the above-mentioned magnesium compounds having reducing ability or compounds derived in the course of preparation of the catalyst component. In order to derive the magnesium compounds having reducing

ability from the magnesium compounds having no reducing ability, for example, the magnesium compounds having reducing ability may be brought into contact with polysiloxane compounds, halogen-containing silane compounds, halogen-containing aluminium compounds, esters, alcohols, halogen-containing compounds, or compounds having a hydroxyl group or an active carbon-oxygen bond.

In addition, the magnesium compound having reducing ability and the magnesium compound having no reducing ability may form complex compounds and double compounds together with other metals such as aluminium, zinc, boron, beryllium, sodium, or potassium, or may also form a mixture with other metallic compounds. The magnesium compound may be used alone or in combination of two or more kinds of the above mentioned compounds.

[0014]

Among the above-mentioned magnesium compounds, the solid magnesium compound can be made to be a liquid-phase compound by using an electron donor (i). The electron donor (i) includes oxygen-containing electron donors such as alcohols, esters, ethers, phenols, ketones, aldehydes, carboxylic acids, organic acid halides, acid amides, acid anhydrides, and alkoxysilanes; and nitrogen-containing electron donors such as ammonias, amines, nitriles,

pyridines, or isocyanates.

[0015]

Specifically, the alcohols include alcohols having 1 to 18 carbon atoms such as methanol, ethanol, propanol, butanol, pentanol, hexanol, 2-ethyl hexanol, octanol, dodecanol, octadecyl alcohol, oleyl alcohol, benzyl alcohol, phenylethyl alcohol, cumyl alcohol, isopropyl alcohol, and isopropylbenzyl alcohol; halogen-containing alcohols having 1 to 18 carbon atoms such as trichloromethanol, trichloroethanol, and trichlorohexanol; and alkoxy alcohols such as 2-propoxy ethanol, 2-butoxy ethanol, 2-ethoxy propanol, 3-ethoxy propanol, 1-methoxy butanol, 2-methoxy butanol, and 2-ethoxy butanol.

[0016]

The esters include organic acid esters having 2 to 18 carbon atoms such as methyl formate, methyl acetate, ethyl acetate, vinyl acetate, propyl acetate, octyl acetate, cyclohexyl acetate, ethyl propionate, methyl butyrate, ethyl valerate, methyl chloroacetate, ethyl dichloroacetate, methyl metacrylate, ethyl crotonate, ethyl cyclohexane carboxylate, methyl benzoate, ethyl benzoate, propyl benzoate, butyl benzoate, octyl benzoate, cyclohexyl benzoate, phenyl benzoate, benzyl benzoate, methyl toluate, ethyl toluate, amyl toluate, ethyl ethylbenzoate, methyl

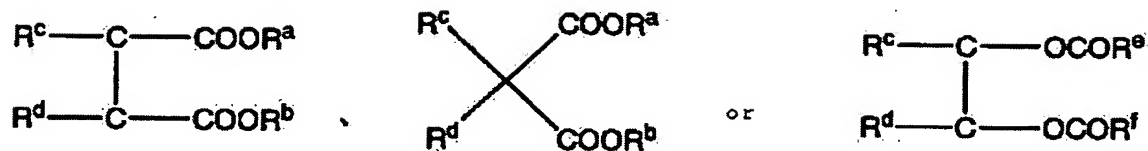
anisate, ethyl anisate, ethyl ethoxybenzoate,  $\gamma$ -butyrolactone,  $\delta$ -valerolactone, cumarine, phthalide, and ethyl carbonate; metallic acid esters such as titanitic acid ester, vanadic acid ester, niobic acid ester, and zirconic acid ester; orthotitanic acid esters such as methyl orthotitanate, ethyl orthotitanate, n-propyl orthotitanate, i-propyl orthotitanate, n-butyl orthotitanate, i-butyl orthotitanate, n-amyl orthotitanate, 2-ethylhexyl orthotitanate, n-octyl orthotitanate, phenyl orthotitanate, and cyclohexyl orthotitanate; polytitanic acid esters such as methyl polytitanate, ethyl polytitanate, n-propyl polytitanate, i-propyl polytitanate, n-butyl polytitanate, i-butyl polytitanate, n-amyl polytitanate, 2-ethylhexyl polytitanate, n-octyl polytitanate, phenyl polytitanate, and cyclohexyl polytitanate; and vanadic acid ester, niobic acid ester, and zirconic acid ester, which are obtained by substituting vanadium, niobium, or zirconium for titanium of titanitic acid ester.

[0017]

In addition, the esters include polycarboxylic acid esters having a skeleton represented by the following formula.

[0018]

[Chemical Formula 5]



(wherein  $R^a$  is a substituted or unsubstituted hydrocarbon group;  $R^b$ ,  $R^e$ , and  $R^f$  are a hydrogen atom, or a substituted or unsubstituted hydrocarbon group;  $R^c$  and  $R^d$  are a hydrogen atom or a substituted or unsubstituted hydrocarbon group, which preferably at least one thereof is a substituted or unsubstituted hydrocarbon group, and  $R^c$  and  $R^d$  may be linked together to form a cyclic structure; when hydrocarbon groups of  $R^a$  to  $R^f$  is substituted, the substituents contain a hetero atom such as nitrogen, oxygen, or sulfur, for example, a group such as C-O-C, COOR, COOH, OH, SO<sub>3</sub>H, -C-N-C-, and NH<sub>2</sub>).

These polycarboxylic acid esters include, in particular, aliphatic polycarboxylic acid ester, alicyclic polycarboxylic acid ester, aromatic polycarboxylic acid ester, and heterocyclic polycarboxylic acid ester.

[0019]

The preferable specific examples of polycarboxylic acid ester having a skeleton represented by the above general formula include diethyl succinate, dibutyl succinate, diethyl methylsuccinate, diaryl methylsuccinate, diisobutyl  $\alpha$ -methylglutarate, diisopropyl  $\beta$ -methylglutarate, diisobutyl



methoxymalonate, dibutyl ethoxymalonate, diethyl ethoxymalonate, diethyl isopropoxymalonate, dibutyl isopropoxymalonate, dibutyl butoxymalonate, dibutyl phenoxymalonate, diethyl diethoxymalonate, dibutyl dibutoxymalonate, diethyl dibutoxymalonate, n-butyl maleate, dibutyl methoxymaleate, dibutyl butoxymaleate, di-2-ethylhexyl fumarate, di-n-hexyl cyclohexenecarboxylate, diethyl nadate, diisopropyl tetrahydrophthalate, diethyl phthalate, monoethyl phthalate, dipropyl phthalate, diisobutyl phthalate, diisopropyl phthalate, ethylisobutyl phthalate, di-n-butyl phthalate, di-n-heptyl phthalate, di-n-octyl phthalate, di-2-ethylhexyl phthalate, di(2-methylpentyl) phthalate, di(3-methylpentyl) phthalate, di(4-methylpentyl) phthalate, di(2,3-dimethylbutyl) phthalate, di(3-methylhexyl) phthalate, di(4-methylhexyl) phthalate, di(5-methylhexyl) phthalate, di(3-ethylpentyl) phthalate, di(3,4-dimethylpentyl) phthalate, di(2,4-dimethylpentyl) phthalate, di(2-methylhexyl) phthalate, di(2-methyloctyl) phthalate, didecyl phthalate, diphenyl phthalate, mixtures of these phthalate diesters, diethyl naphthalene dicarboxylate, dibutyl naphthalene dicarboxylate, triethyl trimellitate, tributyl trimellitate, dibutyl 3,4-furandicarboxylate, diethyl adipinate, dibutyl adipinate, dioctyl sebacinate, dibutyl sebacinate, and the like.

[0020]

Especially preferable examples of the polycarboxylic acid ester include n-butyl maleate, diisobutyl methylmalonate, di-n-hexyl cyclohexenecarboxylate, diethyl nadate, diisopropyl tetrahydrophthalate, diethyl phthalate, diisobutyl phthalate, di-n-butyl phthalate, di-2-ethylhexyl phthalate, dibutyl 3,4-furandicarboxylate, and the like.

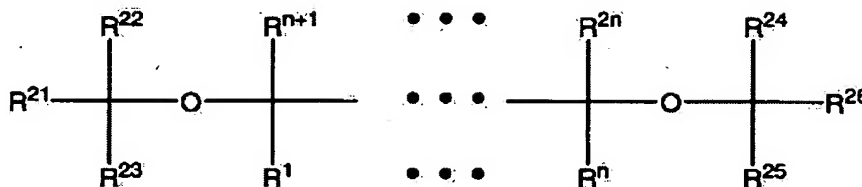
[0021]

The ethers include ethers having 2 to 20 carbon atoms such as methyl ether, ethyl ether, isopropyl ether, butyl ether, amyl ether, tetrahydrofuran, anisol, and diphenylether. In addition, the ethers include a compound having two or more ether linkages with a plurality of bonding atoms existing in the middle (hereinafter, referred as to 'polyether compound'). The polyether compound includes a compound having atoms which exist between the ether linkages and are composed of two or more kinds of atoms selected from carbon, silicon, oxygen, nitrogen, phosphorus, boron, and sulfur. Of these compounds, preferred are a compound in which relatively bulky substituents attaches to the atoms existing between the ether linkages and a plurality of carbon atoms is contained in the atoms existing between two or more ether linkages. For example, there is mentioned a polyether represented by

the following formula.

[0022]

[Chemical Formula 6]



(wherein  $n$  is an integer of  $2 \leq n \leq 10$ ,  $R^1$  to  $R^{26}$  are a substituent having at least one atom selected from carbon, hydrogen, halogen, nitrogen, sulfur, phosphorus, boron and silicon; any of  $R^1$  to  $R^{26}$ , preferably  $R^1$  to  $R^{20}$  may form together a ring other than a benzene ring; and the main chain of the compound may contain atoms other than carbon).

The polyether compound includes 2-(2-ethylhexyl)-1,3-dimethoxypropane, 2-isopropyl-1,3-dimethoxypropane, 2-butyl-1,3-dimethoxypropane, 2-s-butyl-1,3-dimethoxypropane; 2-cyclohexyl-1,3-dimethoxypropane, 2-phenyl-1,3-dimethoxypropane, 2-cumyl-1,3-dimethoxypropane, 2-(2-phenylethyl)-1,3-dimethoxypropane, 2-(2-cyclohexylethyl)-1,3-dimethoxypropane, 2-(p-chlorophenyl)-1,3-dimethoxypropane, 2-(diphenylmethyl)-1,3-dimethoxypropane, 2-(1-naphthyl)-1,3-dimethoxypropane, 2-(2-fluorophenyl)-1,3-dimethoxypropane, 2-(1-decahydronaphthyl)-1,3-dimethoxypropane, 2-(p-t-butylphenyl)-1,3-dimethoxypropane,

2,2-dicyclohexyl-1,3-dimethoxypropane, 2,2-diethyl-1,3-dimethoxypropane, 2,2-dipropyl-1,3-dimethoxypropane, 2,2-dibutyl-1,3-dimethoxypropane, 2-methyl-2-propyl-1,3-dimethoxypropane, 2-methyl-2-benzyl-1,3-dimethoxypropane, 2-methyl-2-ethyl-1,3-dimethoxypropane, 2-methyl-2-isopropyl-1,3-dimethoxypropane, 2-methyl-2-phenyl-1,3-dimethoxypropane, 2-methyl-2-cyclohexyl-1,3-dimethoxypropane, 2,2-bis(p-chlorophenyl)-1,3-dimethoxypropane, 2,2-bis(2-cyclohexylethyl)-1,3-dimethoxypropane, 2-methyl-2-isobutyl-1,3-dimethoxypropane, 2-methyl-2-(2-ethylhexyl)-1,3-dimethoxypropane, 2,2-diisobutyl-1,3-dimethoxypropane, 2,2-diphenyl-1,3-dimethoxypropane, 2,2-dibenzyl-1,3-dimethoxypropane, 2,2-bis(cyclohexylmethyl)-1,3-dimethoxypropane, 2,2-diisobutyl-1,3-diethoxypropane, 2,2-diisobutyl-1,3-dibutoxypropane, 2-isobutyl-2-isopropyl-1,3-dimethoxypropane, 2,2-di-s-butyl-1,3-dimethoxypropane, 2,2-di-t-butyl-1,3-dimethoxypropane, 2,2-dineopentyl-1,3-dimethoxypropane, 2-isopropyl-2-isopentyl-1,3-dimethoxypropane, 2-phenyl-2-benzyl-1,3-dimethoxypropane, 2-cyclohexyl-2-cyclohexylmethyl-1,3-dimethoxypropane, 2,3-diphenyl-1,4-diethoxybutane, 2,3-dicyclohexyl-1,4-diethoxybutane, 2,2-dibenzyl-1,4-diethoxybutane, 2,3-dicyclohexyl-1,4-diethoxybutane, 2,3-diisopropyl-1,4-diethoxybutane, 2,2-bis(p-methylphenyl)-1,4-dimethoxybutane,

2,3-bis(p-chlorophenyl)-1,4-dimethoxybutane, 2,3-bis(p-fluorophenyl)-1,4-dimethoxybutane, 2,4-diphenyl-1,5-dimethoxypentane, 2,5-diphenyl-1,5-dimethoxyhexane, 2,4-diisopropyl-1,5-dimethoxypentane, 2,4-diisobutyl-1,5-dimethoxypentane, 2,4-diisoamyl-1,5-dimethoxypentane, 3-methoxymethyltetrahydrofuran, 3-methoxymethyldioxane, 1,2-diisobutoxypropane, 1,2-diisobutoxyethane, 1,3-diisoamyloxyethane, 1,3-diisoamyloxypropane, 1,3-diisoneopentyloxyethane, 1,3-dineopentyloxypropane, 2,2-tetramethylene-1,3-dimethoxypropane, 2,2-pentamethylene-1,3-dimethoxypropane, 2,2-hexamethylene-1,3-dimethoxypropane, 1,2-bis(methoxymethyl)cyclohexane, 2,8-dioxaspiro[5,5]undecane, 3,7-dioxabicyclo[3,3,1]nonane, 3,7-dioxabicyclo[3,3,0]octane, 3,3-diisobutyl-1,5-oxononane, 6,6-diisobutyldioxyheptane, 1,1-dimethoxymethylcyclopentane, 1,1-bis(dimethoxymethyl)cyclohexane, 1,1-bis(methoxymethyl)bicyclo[2,2,1]heptane, 1,1-dimethoxymethylcyclopentane, 2-methyl-2-methoxymethyl-1,3-dimethoxypropane, 2-cyclohexyl-2-methoxymethyl-1,3-diethoxypropane, 2-cyclohexyl-2-methoxyethyl-1,3-dimethoxypropane, 2,2-diisobutyl-1,3-dimethoxycyclohexane, 2-isopropyl-2-isoamyl-1,3-dimethoxycyclohexane, 2-cyclohexyl-2-methoxymethyl-1,3-dimethoxycyclohexane, 2-isopropyl-2-methoxymethyl-1,3-dimethoxycyclohexane, 2-

isobutyl-2-methoxymethyl-1,3-dimethoxycyclohexane, 2-cyclohexyl-2-ethoxymethyl-1,3-diethoxycyclohexane, 2-cyclohexyl-2-ethoxymethyl-1,3-dimethoxycyclohexane, 2-isopropyl-2-ethoxymethyl-1,3-diethoxycyclohexane, 2-isopropyl-2-ethoxymethyl-1,3-dimethoxycyclohexane, 2-isobutyl-2-ethoxymethyl-1,3-diethoxycyclohexane, and 2-isobutyl-2-ethoxymethyl-1,3-dimethoxycyclohexane.

[0023]

In addition, the polyether includes tris(p-methoxyphenyl)phosphine, methylphenylbis(methoxymethyl)silane, diphenylbis(methoxymethyl)silane, methylcyclohexylbis(methoxymethyl)silane, di-t-butylbis(methoxymethyl)silane, cyclohexyl-t-butylbis(methoxymethyl)silane, i-propyl-t-butylbis(methoxymethyl)silane and the like.

Among these polyether compounds, preferred are 1,3-diethers, especially, 2,2-diisobutyl-1,3-dimethoxypropane, 2-isopropyl-2-isopentyl-1,3-dimethoxypropane, 2,2-dicyclohexyl-1,3-dimethoxypropane, 2,2-bis(cyclohexylmethyl)-1,3-dimethoxypropane, 2-isopropyl-2-cyclohexyl-1,3-dimethoxypropane, 2-isopropyl-2-s-butyl-1,3-dimethoxypropane, 2,2-diphenyl-1,3-dimethoxypropane, and 2-isopropyl-2-cyclopentyl-1,3-dimethoxypropane.

[0024]

The phenols include those having 6 to 20 carbon atoms which may have lower alkyl groups such as phenol, cresol, xylenol, ethylphenol, propylphenol, nonylphenol, cumylphenol, and naphthol. The ketones include those having 3 to 15 carbon atoms such as acetone, methylethylketone, methylisobutylketone, acetophenone, benzophenone, and benzoquinone.

The aldehydes include those having 2 to 15 carbon atoms such as acetaldehyde, propionaldehyde, octylaldehyde, benzaldehyde, tolylaldehyde, and naphthoaldehyde. The organic acid halides include those having 2 to 15 carbon atoms such as acetyl chloride, benzoyl chloride, toluic acid chloride, and anisic acid chloride.

The acid amides include N,N-dimethylacetamide, N,N-diethylbenzamide, N,N-dimethyltoluamide, and the like. The acid anhydrides include acetic anhydride, phthalic anhydride, benzoic anhydride, and the like. The amines include trimethylamine, triethylamine, tributylamine, tribenzylamine, tetramethylethylenediamine, and the like.

The pyridines include pyridine, methyl pyridine, ethyl pyridine, dimethyl pyridine, and the like. These electron donors (i) can be used alone or in combination of two or more kinds. Among these, particularly preferred are

alcohols, alkoxyalcohols, and metallic acid esters. The solubilization reaction of the solid magnesium compound by an electron donor (i) is generally performed by a method in which the solid magnesium compound is brought into contact with the electron donor (i) and be optionally heated. At this time, the contact temperature is 0 to 200 °C, preferably 20 to 180 °C, and more preferably 50 to 150 °C.

In the solubilization reaction, the reaction mixture may coexist with a hydrocarbon solvent or the like. As the hydrocarbon solvents, there can be used, especially, aliphatic hydrocarbons such as pentane, hexane, heptane, octane, decane, dodecane, tetradecane, and petroleum; alicyclic hydrocarbons such as cyclopentane, methylcyclopentane, cyclohexane, methylcyclohexane, cyclooctane, and cyclohexene; aromatic hydrocarbons such as benzene, toluene, and xylene; and halogenated hydrocarbons such as dichloroethane, dichloropropane, trichloroethylene, chlorobenzene, and 2,4-dichlorotoluene.

[0025]

As the magnesium compound used in preparation of the solid titanium catalyst component (a), there can be used many magnesium compounds other than the above-described compounds, but it is preferable that the magnesium compounds exist in the form of a halogen-containing magnesium compound



in the solid titanium catalyst component (a) finally obtained. Therefore, when a magnesium compound containing no halogen is used, it is preferable that the magnesium compound is brought into contact with a halogen-containing compound in the course of preparation of the catalyst component. Among these compounds, the magnesium compounds having no reduction ability are preferred, and the magnesium compounds containing halogen are particularly preferred. Among them, more preferred are magnesium chloride, alkoxy magnesium chloride, aryloxy magnesium chloride, and the like.

[0026]

As the titanium compound used in the preparation of the solid titanium component (a), the tetravalent titanium compound is preferably used, and includes a compound represented by the following formula:



(wherein R is a hydrocarbon group, X is a halogen atom, and  $0 \leq g \leq 4$ ). Examples of such compounds include titanium tetrahalides such as  $\text{TiCl}_4$ ,  $\text{TiBr}_4$ , and  $\text{TiI}_4$ ; alkoxytitanium trihalides such as  $\text{Ti(OCH}_3\text{)Cl}_3$ ,  $\text{Ti(OC}_2\text{H}_5\text{)Cl}_3$ ,  $\text{Ti(O}_n\text{-C}_4\text{H}_9\text{)Cl}_3$ ,  $\text{Ti(OC}_2\text{H}_5\text{)Br}_3$ , and  $\text{Ti(O-iso-C}_4\text{H}_9\text{)Br}_3$ ; dialkoxytitanium dihalides such as  $\text{Ti(OCH}_3\text{)}_2\text{Cl}_2$ ,  $\text{Ti(OC}_2\text{H}_5\text{)}_2\text{Cl}_2$ ,  $\text{Ti(O}_n\text{-C}_4\text{H}_9\text{)}_2\text{Cl}_2$ , and  $\text{Ti(OC}_2\text{H}_5\text{)}_2\text{Br}_2$ ; trialkoxytitanium monohalides such as  $\text{Ti(OCH}_3\text{)}_3\text{Cl}$ ,  $\text{Ti(OC}_2\text{H}_5\text{)}_3\text{Cl}$ ,  $\text{Ti(O}_n\text{-C}_4\text{H}_9\text{)}_3\text{Cl}$ , and  $\text{Ti(OC}_2\text{H}_5\text{)}_3\text{Br}$ ; and tetraalkoxytitaniums such as  $\text{Ti(OCH}_3\text{)}_4$ ,  $\text{Ti(OC}_2\text{H}_5\text{)}_4$ ,  $\text{Ti(O}_n\text{-C}_4\text{H}_9\text{)}_4$ ,

$\text{Ti}(\text{O-iso-C}_4\text{H}_9)_4$ , and  $\text{Ti}(\text{O}_2\text{-ethylhexyl})_4$ .

Among these compounds, preferred are titanium tetrahalides, and particularly preferred is titanium tetrachloride. These titanium compounds can be used alone or in combination of two or more kinds. Further, these titanium compounds may be used in combination with aromatic hydrocarbons, or may be used after be diluted in hydrocarbon or halogenated hydrocarbon. The solid titanium catalyst component (a) is formed by bring the magnesium compound as described above into contact with the titanium compound as described above and, if necessary, a electron donor (ii).

[0028]

In the preparation of the solid titanium catalyst compound (a), an electron donor (ii) is preferably used. As the electron donor (ii), there can be used acid halides, acid amides, nitrils, acid anhydrides, organic acid esters, and polyethers as described below.

[0029]

In particular, the electron donor (ii) includes acid halides having 2 to 15 carbon atoms, such as acetyl chloride, benzoyl chloride, toluic acid chloride, and anisic acid chloride; acid amides such as N,N-dimethylacetamide, N,N-diethylbenzamide and N,N-dimethyltoluamide; nitriles such as acetonitrile, benzonitrile, and tolylnitrile; acid anhydrides such as acetic anhydride, phthalic anhydride, and

benzoic anhydride; and organic acid esters having 2 to 18 carbon atoms, such as methyl formate, methyl acetate, ethyl acetate, vinyl acetate, propyl acetate, octyl acetate, cyclohexyl acetate, ethyl propionate, methyl butyrate, ethyl valerate, methyl chloroacetate, ethyl dichloroacetate, methyl methacrylate, ethyl crotonate, ethyl cyclohexanecarboxylate, methyl benzoate, ethyl benzoate, propyl benzoate, butyl benzoate, octyl benzoate, cyclohexyl benzoate, phenyl benzoate, benzyl benzoate, methyl toluate, ethyl toluate, amyl toluate, ethyl ethylbenzoate, methyl anisate, ethyl anisate, ethyl ethoxybenzoate,  $\gamma$ -butyrolactone,  $\delta$ -valerolactone, cumarine, phthalide, and ethyl carbonate.

[0030]

In addition, preferable examples of the organic acid esters include polycarboxylic acid esters as described above. As the polycarboxylic acid esters, phthalic acid diesters are preferably used. Examples of the electron donor (ii) include polyethers as described above.

[0031]

As the polyethers, there can be preferably used 2,2-diisobutyl-1,3-dimethoxypropane, 2-isopropyl-2-isobutyl-1,3-dimethoxypropane, 2-isopropyl-2-isopentyl-1,3-dimethoxypropane, 2,2-dicyclohexyl-1,3-dimethoxypropane, 2,2-bis(cyclohexylmethyl)-1,3-dimethoxypropane, and the like.

[0032]

As the electron donor (ii), there can be preferably used organic acid esters and polyethers, and more preferably aromatic diesters and polyethers. The electron donor (ii) as described above can be used in combination of two or more kinds. Further, the electron donor (ii) as exemplified above may be finally contained in the solid titanium component (a). Therefore, in the preparation of the solid titanium component (a), the compounds as exemplified above are not always used, and other compounds which are capable of producing such compounds in the course of the preparation of the solid titanium component (a) may be used. At this time, there can also be used other compounds so that two or more kinds of electron donor (ii) is formed.

[0033]

The electron donor (ii) can be used alone and in combination of two or more kinds.

[0034]

The solid titanium component (a) in the carrier-supported form may also be prepared by using a particulate carrier as described below at the time of bringing a titanium compound, a magnesium compound, and optionally an electron donor (ii) into contact with one another as described above.

[0035]

As the carrier, there may be mentioned inorganic carriers such as  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{CaO}$ ,  $\text{TiO}_2$ ,  $\text{ZnO}$ ,  $\text{Zn}_2\text{O}$ ,  $\text{SnO}_2$ ,  $\text{BaO}$ , and  $\text{ThO}$ , and organic carriers such as styrene/divinylbenzene copolymers. Of these carriers, preferred are  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{ZnO}$ , and  $\text{Zn}_2\text{O}$ .

[0036]

In addition, the above-mentioned component may be contacted in the presence of other reagents such as silicon, phosphorus, and aluminium.

[0037]

[The preparation of the solid titanium catalyst component (a)]

The solid titanium catalyst component (a) can be prepared by bringing the titanium compound, the magnesium compound, and the electron donor (ii) into contact with one another as described above, and can be prepared by using all methods including methods known in the art.

[0038]

The process of preparing the solid titanium component (a) is simply described below by way of several examples.

(1) A process wherein a solution consisting of a magnesium compound, a electron donor (ii), and a hydrocarbon solvent is brought into contact and reaction with a titanium compound, after or simultaneously with precipitating a solid by bringing the solution into contact and reaction with an

organoaluminium compound.

(2) A process wherein a complex consisting of a magnesium compound and electron donor (ii) is brought into contact and reaction with a titanium compound and then the product is brought into contact and reaction with an organoaluminium compound.

(3) A process wherein a product obtained by bringing an organic carrier into contact with an organomagnesium compound is brought into contact and reaction with a titanium compound, and preferably electron donor (ii). The contact product may also be brought into contact and reaction with a halogen-containing compound and/or an organoaluminium compound.

(4) A process wherein the magnesium compound-supported organic or inorganic carrier is obtained from a mixture of an inorganic or organic carrier and the solution containing a magnesium compound, electron donor (ii), and further optionally a hydrocarbon solvent, and then the obtained carrier is brought the obtained carrier into contact with the titanium compound.

(5) A process wherein the solid titanium catalyst component on which magnesium and titanium are supported is obtained by bringing an inorganic or organic carrier into contact with the solution containing a magnesium compound, electron donor (ii), and further optionally a hydrocarbon solvent.

(6) A process wherein an organomagnesium compound in the liquid state is brought into contact and reaction with a halogen-containing compound.

(7) A process wherein an organomagnesium compound in the liquid state is brought into contact and reaction with a halogen-containing compound, and then is brought into contact with a titanium compound.

(8) A process wherein a magnesium compound containing a alkoxy group is brought into contact and reaction with a halogen-containing titanium compound.

(9) A process wherein a complex compound consisting of an electron donor (ii) and a magnesium compound containing a alkoxy group is brought into contact and reaction with a titanium compounds.

(10) A process wherein a complex compound consisting of an electron donor (ii) and a magnesium compound containing a alkoxy group is brought into contact with an organoaluminium compound, and then is brought into contact and reaction with the titanium compound.

(11) A process wherein a magnesium compound, an electron donor (ii), and a titanium compound are brought into contact and reaction with one another in any order. In carrying out this reaction, each reactant may be pretreated with a reaction assistant such as the electron donor (ii) and/or the organoaluminium compound or a halogen-containing silicon

compound.

(12) A process wherein a magnesium compound in the liquid state having no reducing ability is allowed to react with the titanium compound in the liquid state, preferably, in the presence of the electron donor (ai), thereby depositing a solid magnesium - titanium composite.

(13) A process wherein the reaction product obtained in the process (12) is allowed to further react with the titanium compound.

(14) A process wherein the reaction product obtained in the process (11) or (12) is allowed to further react with the electron donor (ii) and the titanium compound.

(15) A process wherein a solid product obtained by pulverizing the magnesium compound, preferably, the electron donor (ii), and the titanium compound is treated with any one of halogen, a halogen compound, and an aromatic hydrocarbon. This process may include a step of pulverizing only the magnesium compound, a complex compound consisting of the magnesium compound and the electron donor (ii), or the magnesium compound and the titanium compound. After pulverization, the solid product obtained may be pretreated with a reaction assistant, and then treated with halogen or the like. The reaction assistant may include an organoaluminium compound, a halogen-containing silicon compound, or the like.



(16) A process wherein after pulverization of the magnesium compound, the pulverization compound is brought into contact and reaction with the titanium compound. It is preferable to use the electron donor (ii) or a reaction assistant at the time of pulverization and/or contact reaction.

(17) A process wherein the compound obtained in any of the processes of (11) to (16) is treated with halogen, a halogen compound, or an aromatic hydrocarbon.

(18) A process wherein the reaction product obtained by the contact and reaction of a metallic oxide, an organomagnesium, and a halogen-containing compound is preferably brought into contact with the electron donor (ii) and titanium compound.

(19) A process wherein a magnesium compound such as magnesium salt of an organic acid, alkoxymagnesium, or aryloxymagnesium is allowed to react with the titanium compound and/or a halogen-containing hydrocarbon, and preferably the electron donor (ii).

(20) A process wherein a hydrocarbon solution containing at least the magnesium compound and alkoxytitanium is brought into contact with the titanium compound and/or the electron donor (ii). In this process, It is preferred that halogen-containing compounds such as a halogen containing silicon compound is allowed to coexist in the reaction mixture.

(21) A process wherein a magnesium compound in the liquid

state having no reducing ability is allowed to react with an organoaluminium compound to deposit a solid magnesium metal (aluminium) composite, and then react with the electron donor (ii) and the titanium compound.

[0039]

The amounts of each component used in the preparation of the solid titanium catalyst component (a) cannot be accurately defined because it varies with processes of preparing thereof. For example, the titanium compound is used in an amount of 0.01 to 1000 moles and preferably 0.1 to 200 moles, based on 1 mole of the magnesium compound. The electron donor (ii) optionally used is used in an amount of 0.01 to 5 moles and preferably 0.1 to 1 mole, based on 1 mole of the magnesium compound.

[0040]

The solid titanium catalyst component (a) obtained as describe above contains magnesium, titanium, and halogen. In this solid titanium component (A), halogen / titanium (atomic ratio) is 2 to 200, and preferably 4 to 100, electron donor (ii) / titanium (molar ratio) is 0.01 to 100, and preferably 0.2 to 10, and magnesium / titanium (atomic ratio) is 1 to 100, and preferably 2 to 50.

[0041]

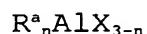
[Organometallic compound catalyst component (b)]

As the organometallic compound catalyst component (b)

used herein, preferred is an organometallic compound catalyst component containing a metal selected from those belonging to Group 13 of the periodic table. Of these, Examples of such compounds include an organoaluminium compound, an organoboron compound, an alkyl complex compound of Group 1 element and aluminium or boron, and the like.

[0042]

The organoaluminium compound can be exemplified by an organoaluminium compound represented by the following formula:



(wherein  $R^a$  is a hydrocarbon group having 1 to 12 carbon atoms, X is halogen or hydrogen, and n is 1 to 3.).

In the above formula,  $R^a$  is a hydrocarbon group having 1 to 12 carbon atoms such as an alkyl group, a cycloalkyl group, or an aryl group, and in particular, a methyl group, an ethyl group, a n-propyl group, an isopropyl group, an isobutyl group, a pentyl group, a hexyl group, an octyl group, a cyclopentyl group, a cyclohexyl group, a phenyl group, or a tolyl group.

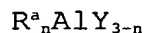
[0043]

The organoaluminium compound described above includes particularly trialkyl aluminiums such as trimethyl aluminium, triethyl aluminium, triisopropyl aluminium, triisobutyl aluminium, trioctylaluminium, and tri-2-ethylhexylaluminium;

trialkenyl aluminium such as triisoprenyl aluminium; dialkylaluminium halides such as dimethyl aluminium chloride, diethyl aluminium chloride, diisopropyl aluminium chloride, diisobutyl aluminium chloride, and dimethyl aluminium bromide; alkyl aluminium sesquihalide such as methyl aluminium sesquichloride, ethyl aluminium sesquichloride, isopropyl aluminium sesquichloride, butylaluminium sesquichloride, ethylaluminium sesquibromide; alkyl aluminium dihalides such as methyl aluminium dichloride, ethyl aluminium dichloride, isopropyl aluminium dichloride, ethyl aluminium dibromide; and alkyl aluminium hydrides such as diethyl aluminium hydride, diisobutyl aluminium hydride, and ethyl aluminium dihydride.

[0044]

As the organoaluminium compound, there can also be used a compound represented by the following formula:



(wherein  $R^a$  has the same meaning as defined above,  $Y$  is  $-OR^b$  group,  $-OSiR^c_3$  group,  $-OAlR^d_2$  group,  $-NR^e_2$  group,  $-SiR^f_3$  group, or  $-N(R^g)AlR^h_2$  group,  $n$  is 1 to 2,  $R^b$ ,  $R^c$ ,  $R^d$  and  $R^h$  are a methyl group, an ethyl group, an isopropyl group, an isobutyl group, a cyclohexyl group, a phenyl group, or the like,  $R^e$  is hydrogen, a methyl group, an ethyl group, an isopropyl group, a phenyl group or a trimethylsilyl group, and  $R^f$  and  $R^g$  are a methyl group or an ethyl group.

[0045]

The organoaluminium compounds include, in particular, compounds described below.

(i) Compounds represented by the formula  $R_n^aAl(OR^b)_{3-n}$  such as dimethyl aluminium methoxide, diethyl aluminium ethoxide, and di-isobutyl aluminium methoxide, (ii) compounds represented by the formula  $R_n^aAl(OSiR^c)_{3-n}$  such as  $Et_2Al(OSiMe_3)$ ,  $(iso-Bu)_2Al(OSiMe_3)$ , and  $(iso-Bu)_2Al(OSiEt_3)$ , (iii) compounds represented by the formula  $R_n^aAl(OAR^d)_{3-n}$  such as  $Et_2AlOAlEt_2$ , and  $(iso-Bu)_2AlOAl(iso-Bu)_2$ , (iv) compounds represented by the formula  $R_n^aAl(NR^e)_{3-n}$  such as  $Me_2AlNEt_2$ ,  $Et_2AlNHMe$ ,  $Me_2AlNHET$ ,  $Et_2AlN(Me_3Si)_2$  and  $(iso-Bu)_2AlN(Me_3Si)_2$ , (v) compounds of the formula  $R_n^aAl(SiR^f)_{3-n}$  such as  $(iso-Bu)_2AlSiMe_3$ , and (vi) compounds represented by the formula  $R_n^aAl[NRa-AlR^h]_{3-n}$  such as  $Et_2AlN(Me)-AlEt_2$  and  $(iso-Bu)_2AlN(Et)Al(iso-Bu)_2$ .

[0046]

Further, compounds analogous to the above compounds, for example, organoaluminium compounds wherein two or more aluminium atoms are linked through an oxygen atom or a nitrogen atom, are also employable. More particularly, examples of such compounds include  $(C_2H_5)_2AlOAl(C_2H_5)_2$ ,  $(C_4H_9)_2AlOAl(C_4H_9)_2$  and  $(C_2H_5)_2AlN(C_2H_5)Al(C_2H_5)_2$ , and in addition, aluminoxanes such as methyl aluminoxane.

[0047]

There can also be used an organoaluminium compound represented by the following formula:



(wherein  $R^a$ , X, and Y have the same meanings as defined above).

The organoboron compound includes triphenylboron, tris(4-fluorophenyl)boron, tris(3,5-difluorophenyl)boron, tris(4-fluoromethylphenyl)boron, tris(pentafluorophenyl)boron, tris(p-tolyl)boron, tris(o-tolyl)boron, tris(3,5-dimethylphenyl)boron, thexylborane, dicyclohexylborane, diamylborane, diisopinocampheylborane, 9-borabicyclo[3,3,1]nonane, catecholborane, B-bromo-9-borabicyclo[3,3,1]nonane, borane-triethylamine complex, and borane-methylsulfide complex.

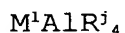
[0048]

Further, as the organoboron compound, there may be used an ionic compound. Examples of such compounds include triethylammonium tetra(phenyl)boron, tripropylammonium tetra(phenyl)boron, trimethylammonium tetra(p-tolyl)boron, trimethylammonium tetra(o-tolyl)boron, tri(n-butyl)ammonium tetra(pentafluorophenyl)boron, tripropylammonium tetra(o,p-dimethylphenyl)boron, tri(n-butyl)ammonium tetra(p-trifluoromethylphenyl)boron, N,N-dimethylanilinium tetra(phenyl)boron, dicyclohexylammonium tetra(phenyl)boron, triphenylcarbenium tetraquis(pentafluorophenyl)borate, N,N-

dimethylanilinium tetraquis(pentafluorophenyl)borate, bis[tri(n-butyl)ammonium]nonaborate, and bis[tri(n-butyl)ammonium]decaborate.

[0049]

As the alkyl complex compound of Group 1 element and aluminium, there can be exemplified a compound represented by the following formula:



(wherein  $M^1$  is Li, Na, or K, and  $R^j$  is a hydrocarbon group having 1 to 15 carbon atoms). In particular, examples of such compound include  $LiAl(C_2H_5)_4$ ,  $LiAl(C_7H_{15})_4$ , and the like.

[0050]

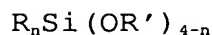
The organoboron compound and the alkyl complex compound of the Group 1 element and boron include those having a structure in which aluminium in the above-described organoaluminium compound and the alkyl complex compound of Group 1 element and aluminium is substituted for boron.

[0051]

The olefin polymerization catalyst may contain the solid titanium catalyst component (a), the organometallic compound catalyst component (ii), and/or an electron donor (iii) described below.

[0052]

The electron donor (iii) includes an organosilicon compound represented by the following general formula:



(wherein R and R' is a hydrocarbon group and is  $0 < n < 4$ ).

Examples of the electron donor (iii) include trimethylmethoxysilane, trimethylethoxysilane, dimethyldimethoxysilane, dimethyldiethoxysilane, diisopropyldimethoxysilane, t-butylmethyldimethoxysilane, t-butylmethyldiethoxysilane, t-amylmethyldiethoxysilane, diphenyldimethoxysilane, phenylmethyldimethoxysilane, diphenyldiethoxysilane, bis-o-tolyldimethoxysilane, bis-m-tolyldimethoxysilane, bis-p-tolyldimethoxysilane, bis-p-tolyldiethoxysilane, bisethylphenyldimethoxysilane, dicyclohexyldimethoxysilane, cyclohexylmethyldimethoxysilane, cyclohexylmethyldiethoxysilane, ethyltrimethoxysilane, ethyltriethoxysilane, vinyltrimethoxysilane, methyltrimethoxysilane, n-propyltriethoxysilane, decyltrimethoxysilane, decyltriethoxysilane, phenyltrimethoxysilane,  $\gamma$ -chloropropyltrimethoxysilane, methyltriethoxysilane, ethyltriethoxysilane, vinyltriethoxysilane, t-butyltriethoxysilane, n-butyltriethoxysilane, iso-butyltriethoxysilane, phenyltriethoxysilane,  $\gamma$ -aminopropyltriethoxysilane, chlorotriethoxysilane, ethyltriisopropoxysilane, vinyltributoxysilane, cyclohexyltrimethoxysilane, cyclohexyltriethoxysilane, 2-norbornanetrimethoxysilane, 2-norbornanetriethoxysilane, 2-norbornanemethyldimethoxysilane,



ethyl silicate, butyl silicate, trimethylphenoxysilane, methyltriallyloxysilane, vinyltris(beta-methoxyethoxysilane), vinyltriacetoxysilane, dimethyltetraethoxydisiloxane, cyclopentyltrimethoxysilane, 2-methylcyclopentyltrimethoxysilane, 2,3-dimethylcyclopentyltrimethoxysilane, cyclopentyltriethoxysilane, dicyclopentyl dimethoxysilane, bis(2-methylcyclopentyl)dimethoxysilane, bis(2,3-dimethylcyclopentyl)dimethoxysilane, dicyclopentyl diethoxysilane, tricyclopentylmethoxysilane, tricyclopentylethoxysilane, dicyclopentylmethylmethoxysilane, dicyclopentylethylmethoxysilane, hexenyltrimethoxysilane, dicyclopentylmethylethoxysilane, cyclopentyl dimethylmethoxysilane, cyclopentyl diethylmethoxysilane, and cyclopentyl dimethylethoxysilane.

[0053]

Among the compounds exemplified above, preferred are ethyltriethoxysilane, n-propyltriethoxysilane, t-butyltriethoxysilane, vinyltriethoxysilane, phenyltriethoxysilane, vinyltributoxysilane, diphenyl dimethoxysilane, phenylmethyl dimethoxysilane, bis-p-tolyl dimethoxysilane, p-tolylmethyl dimethoxysilane, dicyclohexyl dimethoxysilane, cyclohexylmethyl dimethoxysilane, 2-norbornanetriethoxysilane, 2-

norbornanemethyldimethoxysilane, phenyltriethoxysilane, dicyclopentyldimethoxysilane, hexenyltrimethoxysilane, cyclopentyltriethoxysilane, tricyclopentylmethoxysilane and cyclopentyldimethylmethoxysilane.

[0054]

In addition, as the electron donor (iii), there can be used a nitrogen containing electron donor including substituted methylenediamines such as N,N,N',N'-tetramethylmethylenediamine and N,N,N',N'-tetraethylmethylenediamine; and substituted imidazolidines such as 1,3-dibenzylimidazolidine and 1,3-dibenzyl-2-phenylimidazolidine; a phosphorus containing electron donor including phosphorous acid esters such as triethyl phosphite, tri-n-propyl phosphite, triisopropyl phosphite, tri-n-butyl phosphite, triisobutyl phosphite, diethyl-n-butyl phosphite and diethylphenyl phosphate; and an oxygen containing electron donor such as 2,6-substituted tetrahydropyranes and 2,5-substituted tetrahydropyranes. The electron donor (iii) as described above can be used alone or in combination of two or more kinds.

[0055]

[metallocene catalyst]

Next, an example of metallocene catalysts is described. The metallocene catalyst used in the production of polyolefin modified at the terminal thereof is not limited

and includes the metallocene catalyst known in the art. The known metallocene catalyst includes a compound of transition metals such as titanium, vanadium, chrome, zirconium, and hafnium. There can be used the metallocene catalyst in the liquid state or in the solid state under the use condition. In addition, it is unnecessary that the metallocene compound is a single compound. The metallocene compound may be those supported on other compound, or complex compounds or double compounds with other compound.

[0056]

The metallocene compound can be used alone or in combination of two or more kinds, or can be used in combination with the above-described solid titanium catalyst component (a). The metallocene compound can be used in combination with the above-described organometallic compound catalyst component (b). An example of the metallocene catalyst is described below.

[0057]

The metallocene catalyst, for example, consists of (a) a compound of a Group 4 transition metal in the periodic table having a cyclopentadienyl skeleton (hereinafter, referred as to 'metallocene compound (c)'), (d) organoaluminium oxy compound, and if necessary, (e) a particulate carrier.

[0058]

[Metallocene compound (c)]

The metallocene compound is represented by the following formula (1).

$$ML_x \dots (1)$$

In the formula, M is a Group 4 transition metal atom in the periodic table, particularly, zirconium, titanium, or hafnium.

[0059]

L is a ligand coordinated to the transition metal, at least one of L is a ligand having a cyclopentadienyl skeleton, and L other than the ligand having a cyclopentadienyl skeleton is a hydrocarbon group having 1 to 12 carbon atoms, an alkoxy group, an aryloxy group, a trialkylsilyl group, SO<sub>3</sub>R group (provided that R is a hydrocarbon group having 1 to 8 carbon atoms which may have a substituent such as halogen), a halogen atom, or a hydrogen atom, and x is a valence of the transition metal atom.

[0060]

The ligands having a cyclopentadienyl skeleton include, for example, a cyclopentadienyl group, alkyl-substituted cyclopentadienyl groups such as a methylcyclopentadienyl group, a dimethylcyclopentadienyl group, a trimethylcyclopentadienyl group, a tetramethylcyclopentadienyl group, a

pentamethylcyclopentadienyl group, an ethylcyclopentadienyl group, a methylethylcyclopentadienyl group, a propylcyclopentadienyl group, a methylpropylcyclopentadienyl group, a butylcyclopentadienyl group, a methylbutylcyclopentadienyl group, and a hexylpentadienyl group, or an indenyl group, a 4,5,6,7-tetrahydroindenyl group, and a fluorenyl group. These groups may be replaced with a halogen atom or a trialkylsilyl group.

[0061]

In the case where the compound represented by the above-described general formula (1) contains two or more groups having a cyclopentadienyl skeleton, two of the groups having a cyclopentadienyl skeleton may be linked together through an alkylene group such as ethylene or propylene, a substituted alkylene group such as isopropylidene or diphenylmethylenes, a silylene group, or a substituted silylene group such as dimethylsilylene, diphenylsilylene, or methylphenylsilylene.

[0062]

The ligand L other than the ligand having a cyclopentadienyl skeleton may particularly include ligands as described below. The hydrocarbon group having 1 to 12 carbon atoms includes an alkyl group, a cycloalkyl group, an aryl group, and an aralkyl group, and more particularly, the alkyl group includes a methyl group, an ethyl group, a

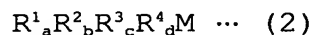
propyl group, an isopropyl group, and a butyl group; the cycloalkyl group includes a cyclopentyl group and a cyclohexyl group; the aryl group includes a phenyl group and a tolyl group; and the aralkyl group includes a benzyl group and a neophyl group.

[0063]

Further, the alkoxy group includes a methoxy group, an ethoxy group, and a butoxy group; the aryloxy group includes phenoxy; and the halogen includes fluorine, chlorine, bromine or iodine; and the ligand represented by  $\text{SO}_3\text{R}$  includes a p-toluenesulfonate group, a methanesulfonate group, or a trifluoromethanesulfonate group.

[0064]

When the valence of the transition metal atom is, for example, 4, the metallocene compound having the cyclopentadienyl skeleton is represented by the following formula (2) in more detail.



In the formula, M is the same transition metal atom as that in the formula (1),  $\text{R}^1$  is a group (ligand) having a cyclopentadienyl skeleton,  $\text{R}^2$ ,  $\text{R}^3$ , and  $\text{R}^4$  are a group having a cyclopentadienyl skeleton, an alkyl group, a cycloalkyl group, an aryl group, an aralkyl group, an alkoxyl group, an aryloxy group, a trialkylsilyl group, a  $\text{SO}_3\text{R}$  group, a halogen atom, or a hydrogen atom, a is an integer of 1 or more, and

$$a + b + c + d = 4.$$

[0065]

The metallocene compound represented by the above-described formula (2) in which at least two of  $R^1$ ,  $R^2$ ,  $R^3$ , and  $R^4$ , for example,  $R^1$  and  $R^2$  are the groups (ligands) having a cyclopentadienyl skeleton is preferably used. The groups having a cyclopentadienyl skeleton may be linked together through an alkylene group such as ethylene or propylene, a substituted alkylene group such as isopropylidene or diphenylmethylene, a silylene group or a substituted silylene group such as dimethylsilylene, diphenylsilylene, or methylphenylsilylene.

[0066]

Further,  $R^3$  and  $R^4$  are a group having a cyclopentadienyl skeleton, an alkyl group, a cycloalkyl group, an aryl group, an aralkyl group, an alkoxyl group, an aryloxy group, a trialkylsilyl group, a  $SO_3R$  group, a halogen atom, or a hydrogen atom. Exemplified below are the metallocene compounds wherein M is zirconium.

[0067]

Bis(indenyl)zirconium dichloride, bis(indenyl)zirconium dibromide, bis(indenyl)zirconium bis(p-toluenesulfonate), bis(4,5,6,7-tetrahydroindenyl)zirconium dichloride, bis(fluorenyl)zirconium dichloride, ethylene bis(indenyl)zirconium dichloride, ethylene

bis(indenyl)zirconium dibromide, ethylene  
 bis(indenyl)dimethylzirconium, ethylene  
 bis(indenyl)diphenylzirconium, ethylene  
 bis(indenyl)methylzirconium monochloride, ethylene  
 bis(indenyl)zirconium bis(methanesulfonate), ethylene  
 bis(indenyl)zirconium bis(p-toluenesulfonate), ethylene  
 bis(indenyl)zirconium bis(trifluoromethanesulfonate),  
 ethylene bis(4,5,6,7-tetrahydroindenyl)zirconium dichloride,  
 isopropylidene(cyclopentadienyl-fluorenyl)zirconium  
 dichloride, isopropylidene(cyclopentadienyl-  
 methylcyclopentadienyl)zirconium dichloride,  
 dimethylsilylene bis(cyclopentadienyl)zirconium dichloride,  
 dimethylsilylene bis(methylcyclopentadienyl)zirconium  
 dichloride,  
 dimethylsilylenebis(dimethylcyclopentadienyl)zirconium  
 dichloride, dimethylsilylenebis(trimethylcyclopentadienyl)  
 zirconium dichloride, dimethylsilylenebis(indenyl)zirconium  
 dichloride, dimethylsilylenebis(indenyl)zirconium  
 bis(trifluoromethanesulfonate), dimethylsilylene bis(4,5,6,7-  
 tetrahydroindenyl)zirconium dichloride,  
 dimethylsilylene(cyclopentadienyl-fluorenyl)zirconium  
 dichloride, diphenylsilylene bis(indenyl)zirconium  
 dichloride, methylphenylsilylene bis(indenyl)zirconium  
 dichloride, bis(cyclopentadienyl)zirconium dichloride,  
 bis(cyclopentadienyl)zirconium dibromide,



bis(cyclopentadienyl)methylzirconium monochloride,  
bis(cyclopentadienyl)ethylzirconium monochloride,  
bis(cyclopentadienyl)cyclohexylzirconium monochloride,  
bis(cyclopentadienyl)phenylzirconium monochloride,  
bis(cyclopentadienyl)benzylzirconium monochloride,  
bis(cyclopentadienyl)zirconium monochloride monohydride,  
bis(cyclopentadienyl)methylzirconium monohydride,  
bis(cyclopentadienyl)dimethylzirconium,  
bis(cyclopentadienyl)diphenylzirconium,  
bis(cyclopentadienyl)dibenzylzirconium,  
bis(cyclopentadienyl)zirconium methoxychloride,  
bis(cyclopentadienyl)zirconium ethoxychloride,  
bis(cyclopentadienyl)zirconium bis(methanesulfonate),  
bis(cyclopentadienyl)zirconium bis(p-toluenesulfonate),  
bis(cyclopentadienyl)zirconium  
bis(trifluoromethanesulfonate),  
bis(methylcyclopentadienyl)zirconium dichloride,  
bis(dimethylcyclopentadienyl)zirconium dichloride,  
bis(dimethylcyclopentadienyl)zirconium ethoxychloride,  
bis(dimethylcyclopentadienyl)zirconium  
bis(trifluoromethanesulfonate),  
bis(ethylcyclopentadienyl)zirconium dichloride,  
bis(methylethylcyclopentadienyl)zirconium dichloride,  
bis(propylcyclopentadienyl)zirconium dichloride,  
bis(methylpropylcyclopentadienyl)zirconium dichloride,

bis(butylcyclopentadienyl)zirconium dichloride,  
 bis(methylbutylcyclopentadienyl)zirconium dichloride,  
 bis(methylbutylcyclopentadienyl)zirconium  
 bis(methanesulfonate),  
 bis(trimethylcyclopentadienyl)zirconium dichloride,  
 bis(tetramethylcyclopentadienyl)zirconium dichloride,  
 bis(pentamethylcyclopentadienyl)zirconium dichloride,  
 bis(hexylcyclopentadienyl)zirconium dichloride, and  
 bis(trimethylsilylcyclopentadienyl)zirconium dichloride.

[0068]

In the compounds exemplified above, the di-substituted cyclopentadienyl ring includes 1,2- and 1,3-substituted compounds, and the tri-substituted cyclopentadienyl ring includes 1,2,3- and 1,2,4-substituted compounds. Further, the alkyl group such as propyl or butyl includes isomer such as n-, iso-, sec-, tert-compounds. The above-described zirconium compounds wherein zirconium has been replaced with titanium or hafnium can also be used as the metallocene compound (c).

[0069]

The metallocene compounds may be used alone, or in combination of two or more kinds. Further, the metallocene compounds may be used after dilution in hydrocarbon or halogenated hydrocarbon. The zirconocene compounds which have zirconium as the central metal atom and have at least

two ligands containing a cyclopentadienyl skeleton are preferably used as the metallocene compound (c).

[0070]

[Organoaluminium oxy compound (d)]

The organoaluminium oxy compound includes the benzene-soluble aluminoxane known in the art or the benzene-insoluble organoaluminium oxy compound as disclosed in Japanese Unexamined Patent Application No. 2-276807. The known aluminoxane can be prepared from (b-2) organoaluminium compounds as described below, for example, by the following processes:

(1) a process of recovering an aluminoxane as its hydrocarbon solution, which comprises adding an organoaluminium compound such as trialkylaluminium to a suspension in a hydrocarbon solvent of a compound containing adsorbed water, or a salt containing water of crystallization such as magnesium chloride hydrate, copper sulfate hydrate, aluminium sulfate hydrate, nickel sulfate hydrate, and cerium chloride hydrate, and then allowing the resulting organoaluminium compound to react with the suspension,

(2) a process of recovering an aluminoxane as its hydrocarbon solution, in which direct water (for example, water, ice, or steam) is allowed to react with an organoaluminium compound such as trialkylaluminium in a

solvent such as benzene, toluene, ethylether and tetrahydrofuran; and

(3) a process for recovering an aluminoxane, in which an organotin oxide such as dimethyltin oxide and dibutyltin oxide is allowed to react with an organoaluminium compound such as trialkylaluminium in a solvent such as decane, benzene, or toluene.

[0071]

Moreover, the solvent or the unreacted organoaluminium compound may be removed from the recovered aluminoxane-containing solution by distillation, and the aluminoxane may be redissolved in a solvent.

[0072]

[Particulate carrier (e)]

As the particulate carrier optionally used in the present invention, there can be mentioned inorganic carriers such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{B}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{ZrO}_2$ ,  $\text{CaO}$ ,  $\text{TiO}_2$ ,  $\text{ZnO}$ ,  $\text{Zn}_2\text{O}$ ,  $\text{SnO}_2$ ,  $\text{BaO}$ , and  $\text{ThO}$ ; and resins (organic carriers) such as polyethylene, polypropylene, poly-1-butene, poly-4-methyl-1-pentene, and styrene/divinylbenzene copolymers. Of these,  $\text{SiO}_2$  is preferable. These can also be used in combination of two or more kinds.

[0073]

When the metallocene catalyst is a solid metallocene catalyst consisting of the metallocene compound (c), the

organoaluminium oxy compound (d), and the particulate carrier (e), the solid metallocene catalyst is formed by supporting the metallocene compound (c) and the organoaluminium oxy compound (d) on the particulate carrier (e) using a method known in the art.

[0074]

Further, the solid metallocene catalyst may be formed by supporting the organoaluminium compound (b-2) as described below together with the metallocene compound (c) and the organoaluminium oxy compound (d) on the particulate carrier (e). In the preparation of the solid metallocene catalyst, the metallocene compound (c) (in terms of a transition metal atom) is usually used in an amount of 0.001 to 1.0 mmole, and preferably 0.01 to 0.5 mmole and the organoaluminium oxy compound (d) is usually used in an amount of 0.1 to 100 mmoles, and preferably 0.5 to 20 mmoles, based on the particulate carrier (e) of 1 g.

[0075]

The solid metallocene catalyst has usually a particle size of 1 to 300  $\mu\text{m}$ , and preferably 10 to 100  $\mu\text{m}$ . In addition, the solid metallocene catalyst may contain other components useful for olefin polymerization such as an electron donor and a reaction assistant together with the catalyst component as described above.

[0076]

In the solid metallocene catalyst used in the present invention, olefin may also be prepolymerized with the solid metallocene catalyst as described above. When olefin polymerization is performed using the prepolymerized metallocene catalyst, the organoaluminium compound (b-2) can also be used together with the above-described metallocene catalyst.

[0077]

Specific examples of the organoaluminium compound (b-2) used as the above-mentioned organoaluminium compound (b-2) or used in the preparation of the above-described organoaluminium oxy compound include trialkylaluminium such as trimethylaluminium, triethylaluminium, tripropylaluminium, triisopropylaluminium, tri-n-butylaluminium, triisobutylaluminium, tri-sec-butylaluminium, tri-tert-butylaluminium, tripentylaluminium, trihexylaluminium, trioctylaluminium, tridecylaluminium, tricyclohexylaluminium, and tricyclooctylaluminium; dialkylaluminium halides such as dimethylaluminium chloride, diethylaluminium chloride, diethylaluminium bromide, and diisobutylaluminium chloride; dialkylaluminium hydrides such as diethylaluminium hydride and diisobutylaluminium hydride; dialkylaluminium alkoxides such as dimethylaluminium methoxide and diethylaluminium ethoxide; and dialkylaluminium aryloxides such as diethylaluminium phenoxide.

[0078]

Among these compounds, trialkylaluminium is preferable, and triethylaluminium and triisobutylaluminium is particularly preferable. Further, as the organoaluminium compound, there can also be used an isoprenylaluminium compound represented by the following general formula:



(wherein x, y, and z are a positive number, and a relation of  $z \geq 2x$ .)

These compounds may also be used in combination of two or more kinds.

[0079]

The organoaluminium compound (b-2) may also contain a small amount of metallic component other than aluminium. When the organoaluminium compound (b-2) together with metallocene compound (c) and organoaluminium oxy compound (d) is supported into the particulate carrier (e), the organoaluminium compound (b-2) is usually used in an amount of 1 to 300 moles, and preferably 2 to 200 moles, based on the solid metallocene catalyst (in terms of transition metal atom) of 1 mole.

[0080]

[Post-metallocene catalyst]

Next, the post-metallocene catalyst is described. The post-metallocene catalyst is a transition metal complex

which contains a nitrogen atom or an oxygen atom in a ligand. The post-metallocene catalyst used in the preparation of polyolefin modified at the terminal thereof is not limited, and the post-metallocene catalyst known in the art is preferably used.

Examples of the post-metallocene catalyst include transition metal complexes described in the following literature.

- 1) M. Brookhart et al., J. Am. Chem. Soc., 117, 6414(1995)
- 2) M. Brookhart et al., J. Am. Chem. Soc., 118, 267(1996)
- 3) D. H. McConville et al., Macromolecules, 29, 5241(1996)
- 4) R. F. Jordan et al., Organometallics, 16, 3282(1997)
- 5) R. H. Grubbs et al., Organometallics, 17, 3149(1998)
- 6) S. Collins et al., Organometallics, 18, 2731(1999)
- 7) M. S. Eisen et al., Organometallics, 17, 3155(1998)
- 8) M. S. Eisen et al., J. Am. Chem. Soc., 120, 8640(1998)
- 9) R. F. Jordan et al. J. Am. Chem. Soc., 119, 8125(1997)
- 10) K. Hakala et al., Macromol. Rapid Commun., 18, 634(1997)

The above-described post-metallocene catalyst may be used alone and in combination of two or more kinds, or may be also used in combination with other olefin polymerization catalyst such as the solid titanium catalyst component (a) or the metallocene compound (d) described above.

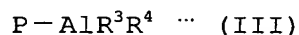
Furthermore, the post-metallocene catalyst can be used in combination with the above-described organometallic compound



catalyst component (b).

[0081]

The polyolefin modified at the terminal thereof represented by the following general formula (III) is produced, for example, in the presence of the olefin polymerization catalyst as described above.



In the formula, P is a polyolefin chain obtained by homopolymerizing or copolymerizing olefins represented by  $CH_2=CHR^2$ .  $R^3$  and  $R^4$  may be the same or different and each is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom, and  $R^2$  is a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom.

[0082]

Specific examples of the olefin represented by  $CH_2=CHR^2$  include ethylene, propylene, butene, pentene, hexene, octene, decene, and the like.

[0083]

Examples of the hydrocarbon group having 1 to 20 carbon atoms include methyl, ethyl, propyl, butyl, pentyl, octyl, decyl, and the like. As P, preferred is a polymer chain having a weight-average molecular weight in the range of 1,000 to 10,000,000. In addition, P is a polymer chain having  $M_w/M_n$  of 1.5 or more, preferably 1.8 or more, and particularly preferably 2 or more.

[0084]

The polyolefin modified at the terminal thereof represented by the general formula (III) is produced by a solvent suspension polymerization method, a suspension polymerization method using olefin in the liquid state as solvent, or the like. When the solvent suspension polymerization is carried out, a hydrocarbon inert to the polymerization reaction can be used as the polymerization solvent. Examples of such inert hydrocarbon solvents include aliphatic hydrocarbons such as propane, butane, pentane, hexane, heptane, octane, decane, dodecane, and petroleum; alicyclic hydrocarbons such as cyclopentane, cyclohexane, and methylcyclopentane; aromatic hydrocarbons such as benzene, toluene, and xylene; halogenated hydrocarbons such as ethylene chloride and chlorobenzene, and a combination thereof. Among these solvents, aliphatic hydrocarbons are particularly preferably used.

[0085]

When the magnesium-supported titanium catalyst system is used, the solid titanium catalyst component (a) or its preliminary polymerization catalyst in the polymerization system is usually used in an amount of about 0.0001 to 50 mmoles, and preferably about 0.001 to 10 mmoles, in terms of titanium atom based on 1 liter of the polymerization volume. The organometallic compound catalyst component (b) is used

in such an amount that the metal atom in the organometallic compound catalyst component (b) is usually in an amount of 1 to 2000 moles, and preferably 2 to 1000 moles, based on 1 mole of titanium atom in the solid titanium catalyst component (a) in the polymerization system. The electron donor (iii) is usually used in an amount of 0.001 to 10 moles, and preferably 0.01 to 5 moles, based on 1 mole of metal atom in the organometallic compound catalyst component (b).

[0086]

When the metallocene catalyst is used as the catalyst, the concentration of the metallocene compound (c) in the polymerization system is usually 0.00005 to 0.1 mmole, and preferably 0.0001 to 0.05 mmole, based on 1 liter of the polymerization volume. The organoaluminium oxy compound (d) is used in an amount of 5 to 1000, and preferably 10 to 400, in terms of molar ratio (Al/M) of an aluminum atom (Al) to a transition metal atom (M) in the metallocene compound (c). When an organoaluminium compound (b-2) is used, it is used usually in an amount of about 1 to 300 moles, and preferably about 2 to 200 moles, based on 1 mole of transition metal atom in the metallocene compound (c).

[0087]

The hydrogen concentration in the polymerization step is 0 to 0.01 mole, preferably 0 to 0.005 mole, and still

more preferably 0 to 0.001 mole, based on 1 mole of the monomer. The olefin polymerization can be carried out by liquid-phase polymerization such as solution polymerization or suspension polymerization or by gaseous-phase polymerization. The reaction solvent used in suspension polymerization can be the above-described inert solvents, or olefins that are liquid at the reaction temperature. The polymerization temperature is usually 70 °C or more, preferably in the range of 80 to 150 °C, preferably in the range of 85 to 140 °C, more preferably 85 to 140 °C, and particularly preferably 90 to 130 °C, and the pressure is usually normal pressure to 10 MPa, and preferably normal pressure to 5 MPa. The polymerization can be carried out in a batch system, a semi-continuous system, or a continuous system, and when the polymerization is carried out in two or more divided stages, the reaction conditions may be the same or different.

[0088]

< Step of converting polyolefin modified at the terminal thereof into polyolefin containing a terminal hydroxyl group  
>

The thus-produced polyolefin modified at the terminal thereof represented by the formula (III) is usually obtained as a slurry. Then, ① the  $-AlR^3R^4$  group in the resulting polyolefin modified at the terminal thereof is subjected to

substitution reaction with a compound having a functional group structure and then subjected to solvolysis, or ② the  $-AlR^3R$  group in the resulting polyolefin modified at the terminal thereof is subjected to substitution reaction with a compound having a structure which forms a functional group by solvolysis and then subjected to solvolysis, to produce polyolefin having a hydroxyl group at the terminal thereof, represented by the following general formula (II).

[0089]

$P-OH \dots (II)$

In the formula (II), P has the same meaning as defined above. The compound having a functional group structure includes a halogen gas, methyl chloroformate, and phthalic acid chloride. The compound having a structure which forms a functional group by solvolysis includes oxygen, carbon monoxide, and carbon dioxide.

[0090]

The substitution reaction of the  $-AlR^3R^4$  group in the resulting polyolefin modified at the terminal thereof with the compound having a functional group structure or the compound having a structure which forms a functional group by solvolysis is usually carried out at a temperature of 0 to 300°C, and preferably 10 to 200 °C, for 0 to 100 hours, and preferably 0.5 to 50 hours. After the substitution reaction is carried out, the solvolysis temperature is

usually a temperature of 0 to 100°C, and preferably 10 to 80 °C, and the solvolysis time is 0 to 100 hours, and preferably 0.5 to 50 hours. The solvent used in solvolysis includes methanol, ethanol, propanol, butanol, water, and the like.

[0091]

The polyolefin having a hydroxyl group at the terminal thereof can be produced by reacting polyolefin having an unsaturated bond at one terminal thereof with an compound containing the group 13 element, for example, an organoaluminium compound or an organoboron compound to form polyolefin modified at one terminal thereof represented by the above described general formula (III), and then converting the terminal into a hydroxyl group in the same manner as described above.

[0092]

The polyolefin having an unsaturated bond at one terminal thereof (also referred to as 'polyolefin unsaturated at one terminal thereof) can be produced by polymerization or copolymerization of olefins having 2 to 20 carbon atoms in the presence of the olefin polymerization catalyst as described above. As the olefin having 2 to 20 carbon atoms, there can be preferably used ethylene, propylene, butane, 3-methyl-1-butene, 3-methyl-1-pentene, 4-methyl-1-pentene, and the like.

[0093]

The olefin polymerization can be carried out by liquid phase polymerization such as solution polymerization or suspension polymerization or by gaseous phase polymerization. The form of polymerization is preferably employed in the reaction form of suspension polymerization. The reaction solvent used in the suspension polymerization can be the above-described inert hydrocarbon solvents, or olefins that are liquid at the reaction temperature.

[0094]

Examples of such inert hydrocarbon solvents include aliphatic hydrocarbons such as propane, butane, pentane, hexane, heptane, octane, decane, dodecane, and petroleum; alicyclic hydrocarbons such as cyclopentane, cyclohexane, and methylcyclopentane; aromatic hydrocarbons such as benzene, toluene, and xylene; halogenated hydrocarbons such as ethylene chloride and chlorobenzene, and a combination thereof. Among these solvents, aliphatic hydrocarbons are particularly preferably used.

[0095]

When polyolefin unsaturated at the terminal thereof is produced by using the magnesium-supported titanium catalyst system as a catalyst, the solid titanium catalyst component (a) or its preliminary polymerization catalyst is used usually in an amount of about 0.001 to 100 mmols, and

preferably about 0.005 to 20 mmoles, in terms of titanium atom based on 1 liter of the polymerization volume. The organometallic compound catalyst component (b) is used in such an amount that the metal atom in the organometallic compound catalyst component (b) is usually in an amount of 1 to 2000 moles, and preferably 2 to 500 moles, based on 1 mole of titanium atom in the solid titanium catalyst component (a) in the polymerization system. The electron donor (iii) is used usually in an amount of 0.001 to 10 moles, and preferably 0.01 to 5 moles, based on 1 mole of metal atom in the organometallic compound catalyst component (b).

[0096]

The hydrogen concentration in the polymerization step is usually 0 to 0.25 mole, preferably 0 to 0.20 mole, and more preferably 0 to 0.15 mole, based on 1 mole of the monomer. The polymerization temperature in the case of using the magnesium-supported titanium catalyst as a catalyst is usually in the range of about 20 to 300 °C, and preferably about 50 to 150 °C, and the polymerization pressure is in the range of 0.01 to 10 MPa, and preferably 0.05 to 5 MPa.

[0097]

When the polyolefin unsaturated at the terminal thereof is produced by using the metallocene catalyst as a catalyst,



the concentration of the metallocene compound (c) in the polymerization system is usually in an amount of 0.00005 to 0.1 mmole, and preferably 0.0001 to 0.005 mmole. The organoaluminium oxy compound (d) is used, in terms of the molar ratio (Al/M) of an aluminium atom (Al) based on a transition metal atom (M), in an amount of 5 to 10000, and preferably 10 to 4000. Further, the organoaluminium compound (b-2) is usually used in an amount of about 1 to 300 moles, and preferably about 2 to 200 moles, based on 1 mole of the transition metal atom in the metallocene compound (c).

[0098]

The polymerization temperature in the case of using the metallocene catalyst is usually in the range of -20 to 180 °C, preferably 0 to 150 °C, and more preferably 0 to 120 °C, and the polymerization pressure is in the range of 0 to 8 MPa, and preferably 0 to 5 MPa. The olefin polymerization can be carried out in a batch system, a semi-continuous system, or a continuous system, and the polymerization can be also carried out in two or more divided stages. In the olefin polymerization, a homopolymer of olefins or a random copolymer of two or more olefins may be produced.

[0099]

The polyolefin unsaturated at the terminal thereof obtained in this manner is reacted with the compound

containing the group 13 element to convert the terminal into a terminal having the group 13 element bound thereto. In addition, when the resulting polyolefin is a mixture of the polyolefin having the group 13 element bound to one terminal thereof and the polyolefin having an unsaturated bond at one terminal thereof, if necessary, the terminal of polyolefin having unsaturated bond at the one terminal can be converted into a terminal having the group 13 element bound thereto.

[0100]

The compound containing the group 13 element used in the reaction can be selected from the compounds exemplified as the organometallic compound catalyst component (b). Preferably, an organoaluminium compound or an organoboron compound can be used. Among those compounds, more preferably is used trialkylaluminum, dialkylaluminum hydride, or a boron compound having one or more hydrogen-boron bonds. The organoaluminium is particularly preferably dialkyl aluminum hydride, and the organoboron compound is particularly preferably 9-borabicyclo[3,3,1]nonane.

[0101]

The reaction of the polyolefin unsaturated at one terminal thereof with the compound containing the group 13 element is carried out, for example, in the following manner

① 0.1 to 50 g of polypropylene having a vinylidene group at the terminal and 5 to 1000 ml of 0.01 to 5 moles/L

diisobutylaluminum hydride in octane are mixed, and are then refluxed for 0.5 to 6 hours.

② 0.1 to 50 g of polypropylene having a vinylidene group at the terminal, 5 to 1000 ml of anhydrous tetrahydrofuran, and 0.1 to 50 ml of 0.05 to 10 moles/L 9-borabicyclo[3,3,1]nonane in tetrahydrofuran are mixed, and are then stirred at 20 to 65 °C for 0.5 to 24 hours.

[0102]

In the above-described manner, the polyolefin modified at one terminal thereof is produced. The group 13 element is bound to the obtained one terminal of polyolefin, and the group 13 element bound above is preferably aluminium.

[0103]

The production of polyolefin having an acryloyl group and a methacryloyl group at the terminal thereof

The polyolefin macromonomer (MM) having an acryloyl group or a methacryloyl group at the terminal thereof represented by the formula (I) is obtained by allowing the polyolefin having a hydroxyl group at the terminal thereof represented by the formula (II) to react with acryloyl halide, methacryloyl halide, acrylic acid, or methacrylic acid.

[0104]

The reaction of the polyolefin having a hydroxyl group at the terminal thereof with acryloyl halide, methacryloyl

halide, acrylic acid, or methacrylic acid can be carried out, for example, in the following manner:

① a process of reacting the polyolefin having a hydroxyl group at the terminal thereof with acryloyl halide or methacryloyl halide such as acrylic acid chloride or methacrylic acid chloride in the presence of a base such as triethylamine, or

② a process of allowing the polyolefin having a hydroxyl group at the terminal thereof to react with acrylic acid or methacrylic acid in the presence of an acid catalyst.

[0105]

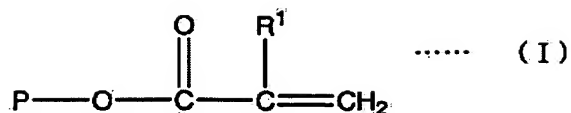
In the reaction, acryloyl halide, methacryloyl halide, acrylic acid, or methacrylic acid is used in the range of 0.1 to 1000 moles, preferably 0.2 to 500 moles, based on 1 mole of the hydroxyl group at the terminal of the polyolefin. The reaction temperature is usually -100 to 150 °C, and preferably 0 to 120 °C, and the reaction time is usually 0.1 to 48 hours, and preferably 0.5 to 12 hours.

[0106]

Therefore, the polyolefin having an acryloyl group or a methacryloyl group at the terminal thereof is produced.

[0107]

[Chemical Formula 7]



In the formula (1-a),  $\text{R}^1$  is an hydrogen atom and a methyl group, and P is a polyolefin chain obtained by homopolymerizing or copolymerizing olefins represented by  $\text{CH}_2=\text{CHR}^2$  (where  $\text{R}^2$  is one selected from a hydrocarbon group having 1 to 20 carbon atoms, a hydrogen atom, or a halogen atom.). In addition, the polyolefin having an acryloyl group or a methacryloyl group at the terminal thereof has  $\text{Mw/Mn}$  of 1.5 or more, preferably 1.8 or more, and particularly preferably 2 or more.

[0108]

[Examples]

Hereinafter, The present invention will be described in more detail with reference to the following Examples, however, the present invention is not limited to these examples.

< Example 1 >

[Synthesis of an ethylene/propylene random copolymer modified at the terminal thereof]

800 ml of purified toluene was introduced into a glass autoclave having an internal volume of 1 liter purged sufficiently with nitrogen, and the liquid phase and gaseous phase were saturated by blowing 10 liters/h of ethylene and

90 liters/h of propylene. Thereafter, MAO in an amount of 10 mmole in terms of Al and 0.01 mmole of dicyclopentadienyl zirconiumdichloride were added at 50 °C to initiate polymerization. After polymerization under normal pressure at 50 °C for 120 minutes, a small amount of isobutylalcohol was added to terminate the polymerization. The reaction solution was washed 3 times with 300 ml of 1 N aqueous hydrochloric acid and further 2 times with 200 ml of water, and then the organic layer was dried over anhydrous magnesium sulfate and filtered through a glass filter (G3) to remove the magnesium sulfate. The filtrate was concentrated, and the resulting oily matter was dried under vacuum for 10 hours to obtain 140.4 g colorless transparent oily ethylene/propylene random copolymer (hereinafter, referred as to 'EPR'). When the molecular weight (in terms of EPR) of the polymer was determined by GPC, the  $M_w$  was 2170, the  $M_n$  was 520, and the  $M_w/M_n$  ratio was 4.2. IR analysis indicated that the content of propylene in the polymer was 48 mol%, and 19.6 terminal vinylidene groups were contained per 1000 carbon atoms. 50 g of the resulting EPR containing terminal vinylidene groups were introduced into a 500 ml glass reactor purged sufficiently with nitrogen, and 250 ml of toluene and 50 ml of diisobutyl aluminum hydride were added thereto, and the mixture was heated at 110 °C for 6 hours under stirring. A toluene

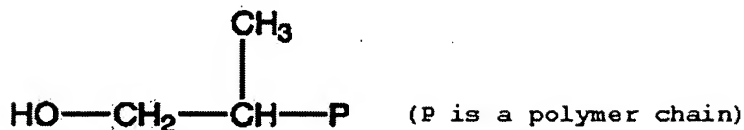
solution containing the EPR modified at the terminal thereof was obtained in this manner.

[Synthesis of EPR containing terminal hydroxyl groups]

While the toluene solution obtained above was kept at 110 °C, a nitrogen gas was replaced with dry air, and while this temperature was kept, the dry air was continuously supplied thereto at a flow rate of 100 liters/h for 6 hours. Thereafter, the solution was transferred to a separatory funnel, washed 3 times with 300 ml of 1 N aqueous hydrochloric acid and then further 3 times with 300 ml of water. The organic layer was dried with anhydrous magnesium sulfate and filtered through a glass filter (G3), and then the filtrate was concentrated. The resulting yellow oily matter was dried for 10 hours under vacuum to obtain 47.0 g of oily polymer. When the molecular weight (in terms of EPR) of the polymer was determined by GPC, the  $M_w$  was 2200, the  $M_n$  was 520, and the  $M_w/M_n$  ratio was 4.3. A sample obtained by dissolving 100 mg polymer in 0.6 ml chloroform- $d$  at 25 °C was analyzed with  $^1H$ -NMR (JEOL GSX-270 manufactured by JEOL. Ltd.). As a result, it indicated a signal at 3.5 ppm attributable to a methylene group adjacent to a hydroxyl group. That is, the presence of EPR having a terminal structure represented by the structure described below was confirmed. From the integrated value, the content of hydroxyl groups was calculated to be 2.4 mol%.

[0109]

[Chemical Formula 8]



[0110]

[Synthesis of EPR macromonomer]

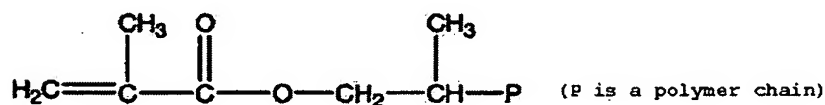
20 g of the ERP having terminal hydroxyl groups obtained above was introduced into a 100 ml Schlenk tube purged sufficiently with nitrogen, and 20 ml of toluene, 2.7 ml of triethylamine and 2.5 ml of methacryloyl chloride were added thereto, and the mixture was stirred at room temperature for 3.5 hours. The resulting reaction solution was transferred to a separatory funnel, washed 3 times with 100 ml of 1 N aqueous hydrochloric acid and then further 3 times with 100 ml of water. The organic layer was dried with anhydrous magnesium sulfate and filtered through a glass filter (G3), and then the filtrate was concentrated. The resulting yellow oily matter was dried for 10 hours under vacuum to obtain 21.1 g of yellow oily polymer. When the molecular weight (in terms of EPR) of the polymer was determined by GPC, the Mw was 2240, the Mn was 690, and the Mw/Mn ratio was 3.3. A sample obtained by dissolving 100 mg of polymer in 0.6 ml of chloroform-d at 25 °C was analyzed with <sup>1</sup>H-NMR (JEOL GSX-270 manufactured by JEOL, Ltd.). As a



result, it indicated signals at 1.95 ppm attributable to a methyl group in a methacryloyl group, at 3.8 to 4.2 ppm attributable to a methylene group adjacent to an ester group, and at 5.5 and 6.1 ppm attributable to a vinyl group in a methacryloyl group. That is, the presence of EPR macromonomer having a terminal structure represented by the structure described below was confirmed. From the integrated value, the content of methacryloyl groups was calculated to be 2.6 mol%, thus revealing almost quantitative conversion of hydroxyl groups in the starting material.

[0111]

[Chemical Formula 9]



[0112]

< Example 2 >

[Synthesis of polyethylene modified at the terminal thereof]

800 ml of purified decane was introduced into a glass autoclave having an internal volume of 1 L purged sufficiently with nitrogen, and the liquid phase and the gaseous phase were saturated by blowing 100 liters/h of ethylene, respectively. Thereafter, MAO in an amount of 20 mmole in terms of Al and 0.02 mmole of dicyclopentadienyl

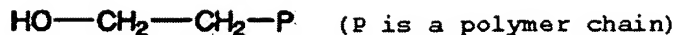
zirconium dichloride were added at 100 °C to initiate polymerization. After polymerization at normal pressure at 100 °C for 120 minutes, a small amount of isobutylalcohol was added to terminate the polymerization. The reaction solution was introduced into a mixed solution of 1.5 L of methanol and 1.5 L of acetone, and a small amount of 1 N aqueous hydrochloric acid was added thereto, and the mixture was stirred. The precipitated polymer was separated by filtration with a glass filter from the aqueous phase and dried at 80 °C for 10 hours under reduced pressure. By the operation as described above, 24.6 g of white polymer was obtained. When the molecular weight (in terms of PE) of the polymer was determined by GPC, the Mw was 15200, the Mn was 5300, and the Mw/Mn ratio was 2.9. IR analysis indicated that 1.1 terminal vinyl groups were contained per 1000 carbon atoms. 14.3 g of the resulting terminal vinyl group-containing polyethylene was introduced into a 1 L glass reactor purged sufficiently with nitrogen, and 750 ml of decane and diisobutyl aluminum hydride in an amount of 6.25 mmole in terms of Al were added thereto, and the mixture was heated at 110 °C for 6 hours while stirring. A decane solution containing polyethylene modified at the terminal thereof was obtained in this manner.

[Synthesis of polyethylene containing terminal hydroxyl groups]

While the decane solution obtained above was kept at 110 °C, a nitrogen gas was replaced with dry air, and while this temperature was maintained, dry air was continuously supplied thereto at a flow rate of 100 liters/h for 6 hours, and the reaction slurry was introduced into a mixed solution of 1.5 L methanol and 1.5 L acetone, and a small amount of 1 N aqueous hydrochloric acid was added thereto and stirred. The precipitated polymer was separated by filtration with a glass filter from the liquid phase and dried at 80 °C for 10 hours under reduced pressure. By the above-mentioned operation, 13.3 g of white polymer was obtained. When the molecular weight (in terms of PE) of the polymer was determined by GPC, the Mw was 15200, the Mn was 5300, and the Mw/Mn ratio was 2.9. A sample obtained by dissolving 100 mg of polymer in 0.6 ml o-dichlorobenzene-d4 at 120 °C was analyzed with <sup>1</sup>H-NMR (JEOL GSX-400 manufactured by JEOL. Ltd.). As a result, it indicated a signal at 3.5 ppm attributable to a methylene group adjacent to a hydroxyl group. That is, the presence of polyethylene having a terminal structure represented by the structure described below was confirmed. From the integrated value, the content of OH groups was calculated to be 0.17 mol%.

[0113]

[Chemical Formula 10]



[0114]

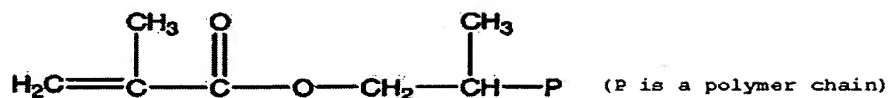
[Synthesis of polyethylene macromonomer]

5.0 g of the thus obtained polyethylene containing terminal hydroxyl groups was introduced into a 100 ml Schlenk tube purged sufficiently with nitrogen, then 20 ml of toluene, 0.7 ml of triethylamine, and 1.0 ml of methacryloyl chloride were added thereto, and the mixture was heated at 80 °C for 3 hours while stirring. The resulting reaction solution was poured into 300 ml of methanol, and a small amount of 1 N aqueous hydrochloric acid was added thereto and stirred. The precipitated polymer was separated by filtration with a glass filter from the liquid phase and dried at 80 °C for 10 hours under reduced pressure. By the above-described operation, 4.9 g of white polymer was obtained. A sample obtained by dissolving 50 mg of polymer in 0.6 ml of o-chlorobenzene-d<sub>4</sub> at 120 °C was analyzed with IR-NMR (JEOL GSX-270 manufactured by JEOL. Ltd.). As a result, it indicated signals at 4.3 ppm attributable to a methylene group adjacent to an ester group and at 5.6 and 6.2 ppm attributable to a vinyl group in a methacryloyl group. That is, the presence of a polyethylene macromonomer having a terminal structure represented by the structure as described

below was confirmed.

[0115]

[Chemical Formula 11]



[0116]

<Example 3>

[Preparation of the solid titanium catalyst component (a)]

95.2 g of anhydrous magnesium chloride, 485 ml of decane, and 390.4 g of 2-ethylhexylalcohol were heated at 140 °C for 4 hours to form a uniform solution, and 22.2 g of phthalic anhydride was added to this solution, and the mixture was mixed at 130 °C for 1 hour while stirring, whereby the phthalic anhydride was dissolved. The thus obtained uniform solution was cooled to room temperature, and 30 ml of the uniform solution was added dropwise over 45 minutes to 80 ml of titanium tetrachloride kept at -20 °C. The temperature of the solution was increased to 110 °C over 4 hours, and when the temperature reached 110 °C, 2.0 ml of diisobutyl phthalate was added thereto, and the mixture was heated at 110 °C for 2 hours. Then, solids were collected by hot filtration, and the solids were suspended again in 110 ml of titanium tetrachloride, and then reacted at 110 °C for 2 hours while heating. After the reaction was finished, the solids were collected again by hot filtration and washed

sufficiently with decane at 110 °C and hexane at room temperature until a free titanium compound was not detected in the solution. The catalyst component (a) prepared by the above-mentioned operation was stored as a hexane slurry, a part of which was dried and examined for the composition, indicating 2.3 wt% of titanium, 19.0 wt% of magnesium and 11.8 wt% of diisobutyl phthalate.

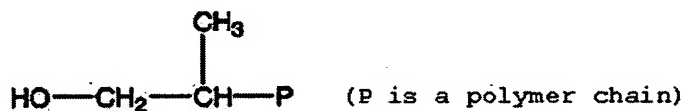
[Synthesis of polypropylene modified at the terminal thereof]

400 ml of purified decane was introduced into a glass autoclave having an internal volume of 500 ml purged sufficiently with nitrogen, and the liquid phase and the gaseous phase were saturated by blowing 100 liters/hr of propylene. Thereafter, 10 mmole of triethylaluminum, 1.0 mmole of cyclohexylmethyldimethoxysilane, and the solid titanium component (a) in an amount of 0.2 mmole in terms of titanium atom were added in this order. The mixture was polymerized at 100 °C for 2 hours while the flow rate of propylene was regulated so that the unreacted gas might leak out through a bubbler tube connected to a purge line and the inside of the reactor was not under reduced pressure. Then, the propylene gas was replaced by a nitrogen gas with which the unreacted propylene gas was purged to terminate the reaction, to obtain decane slurry containing polypropylene modified at the terminal thereof.

[Synthesis of polypropylene containing terminal hydroxyl groups]

The decane slurry obtained above was maintained at 100 °C and reacted for 7 hours while dry air was allowed to pass there through at a flow rate of 200 liters/h. After the reaction was finished, the reaction slurry was introduced into a mixture of 2 L of methanol and 2 L of acetone, and a small amount of 1 N aqueous hydrochloric acid was added thereto and stirred. The precipitated polymer was separated by filtration with a glass filter from the liquid phase and dried at 80 °C for 10 hours under reduced pressure. By the above-mentioned operation, 4.2 g of white polymer was obtained. When the molecular weight (in terms of PP) of the polymer was measured by GPC, the  $M_w$  was 140000, the  $M_n$  was 11100, and the  $M_w/M_n$  ratio was 12.6. By IR analysis, an absorption based on shrinkage vibration of hydroxyl groups which was recognized at  $3640\text{ cm}^{-1}$  was confirmed, and it was confirmed that hydroxyl groups were certainly contained in the polymer. That is, the presence of polypropylene having a structure represented by the structure described below was confirmed.

[Chemical Formula 12]



[0117]

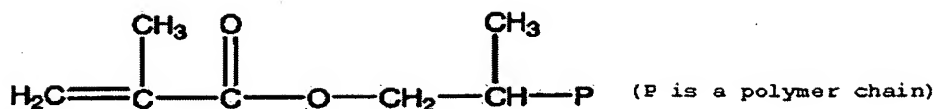
[Synthesis of isotactic polypropylene macromonomer]

1.0 g of the thus obtained polypropylene containing terminal hydroxyl groups was introduced into a 100 ml Schlenk tube purged sufficiently with nitrogen, then 20 ml of toluene, 0.7 ml of triethylamine, and 1.0 ml of methacryloylchloride were added thereto, and the mixture was stirred at 80 °C for 3 hours. The resulting reaction solution was poured into 300 ml of methanol, and a small amount of 1 N aqueous hydrochloric acid was added thereto and stirred. The precipitated polymer was separated by filtration with a glass filter from the liquid phase and dried at 80 °C for 10 hours under reduced pressure. By the above-mentioned operation, 0.9 g of white polymer was obtained. A sample obtained by dissolving 50 mg of the polymer in 0.6 ml of o-chlorobenzene-d<sub>4</sub> at 120 °C was analyzed with <sup>1</sup>H-NMR (JEOL GSX-270 manufactured by JEOL. Ltd.). As a result, it indicated signals at 4.1 to 4.2 ppm attributable to a methylene group adjacent to an ester group and at 5.6 and 6.2 ppm attributable to a vinyl group in a methacryloyl group. That is, the presence of the polypropylene macromonomer having a terminal structure represented by the structure described below was confirmed.

[0118]

[Chemical Formula 13]





[0119]

## [Effect of the Invention]

According to present invention, a novel polyolefin macromonomer can be provided by using an industrially useful method wherein a hydroxyl group is introduced into the terminal of polyolefin produced by using a coordination polymerization catalyst containing a transition metal compound represented by a solid-phase titanium catalyst or a metallocene catalyst used widely in the industry as a catalyst for production of polyolefin, and then a acryloyl group or a methacryloyl group is further introduced into the polyolefin.

[Name of Document] ABSTRACT

[Abstract]

[Object]

The objection of the present invention is to develop a polyolefin macromonomer which consists of various  $\alpha$ -olefin homopolymers or copolymers and is capable of covering a broad molecular weight range, and to provide a process of efficiently producing the macromonomer.

[Solving Means]

A novel polyolefin macromonomer having an acryloyl group and a methacryloyl group at the terminal of polyolefin chain P can be efficiently produced by sequentially carrying out i) a step of producing polyolefin having a hydroxyl group at the terminal of a polyolefin chain and ii) a step of converting a terminal hydroxyl group in the polyolefin chain into an acryloyl group or a methacryloyl group.

[Selected Figure] none